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TITLE: REVIEW OF RADIOACTIVE LIQUID WASTE
MANAGEMENT AT LOS ALAMOS

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REVIEW OF RADIOACTIVE LIQUID WASTE MANAGEMENT
AT LOS ALAMOS

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ABSTRACT - This paper describes in detail the management of radioactive liquid wastes at the Los Alamos Scientific Laboratory, bringing previous reports up-to-date and discussing recent changes and proposed improvements. The wastes originate in a variety of research and development activities and are all in the low-level category. Their management, however, commands a considerable effort because of large volumes and the presence of transuranic radionuclides. Technical concerns have created increasing pressures for reduction of activity levels in discharges with transuranics being of primary importance. Though present treatment facilities are highly efficient, continuing efforts are being made to improve them.

Extensive networks of sewers form the primary collection systems which deliver the wastes to two treatment facilities. Ferric hydroxide precipitation processes at both plants concentrate the alpha radioactivity into sludges which are dewatered at one facility and mixed with cement at the other. The sludges are packaged for storage or burial depending upon whether their transuranic radionuclide content is above or below the 10 nCi/g level.

The paper reviews plans which are in progress to upgrade both collection and treatment systems to meet proposed and expected requirements.

I. INTRODUCTION AND BACKGROUND INFORMATION

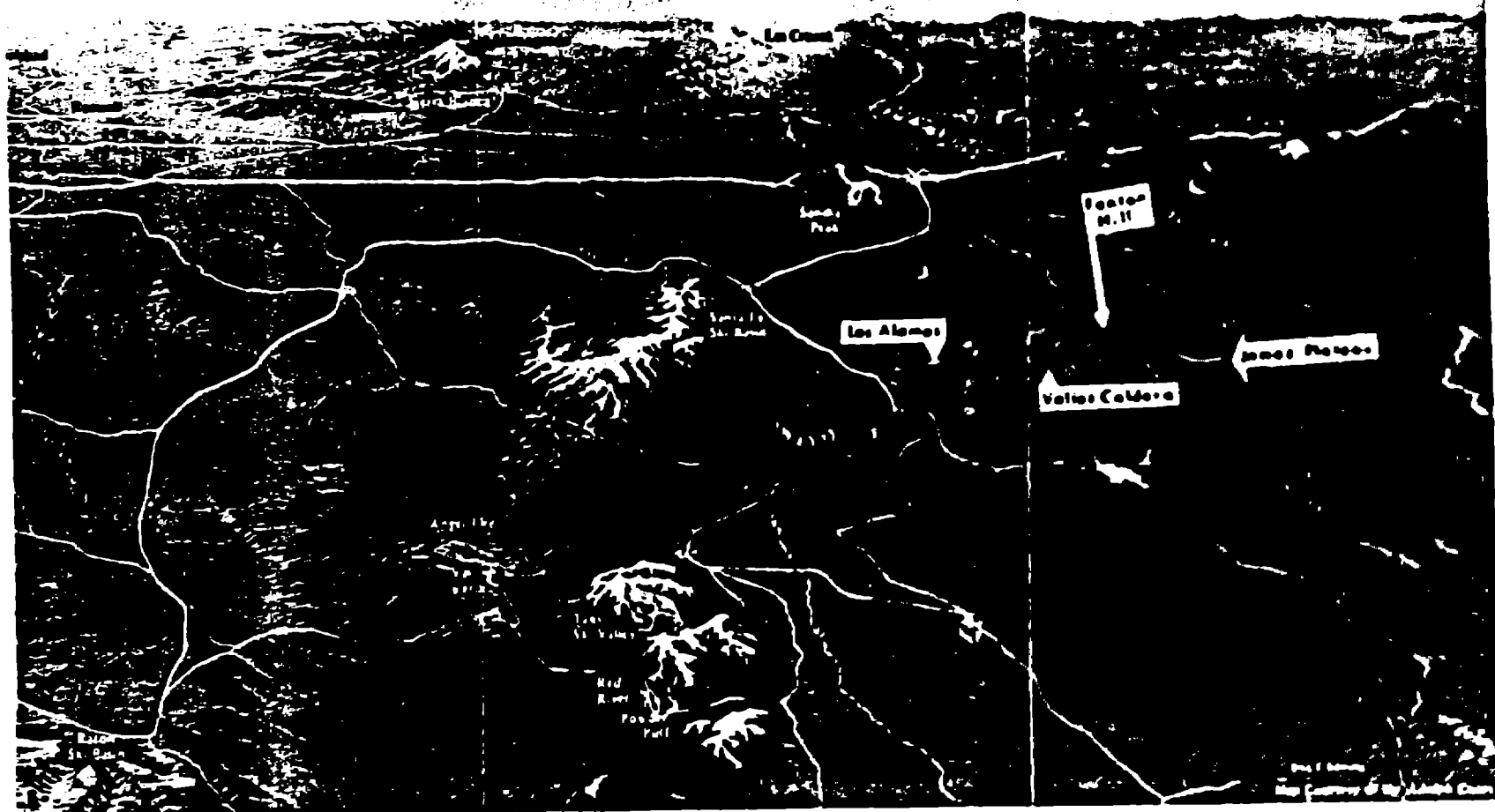
The Los Alamos Scientific Laboratory (LASL) occupies a large portion of Los Alamos County which is located on the Pajarito Plateau of the Jemez Mountains in north central New Mexico. The geology is volcanic in origin with deep

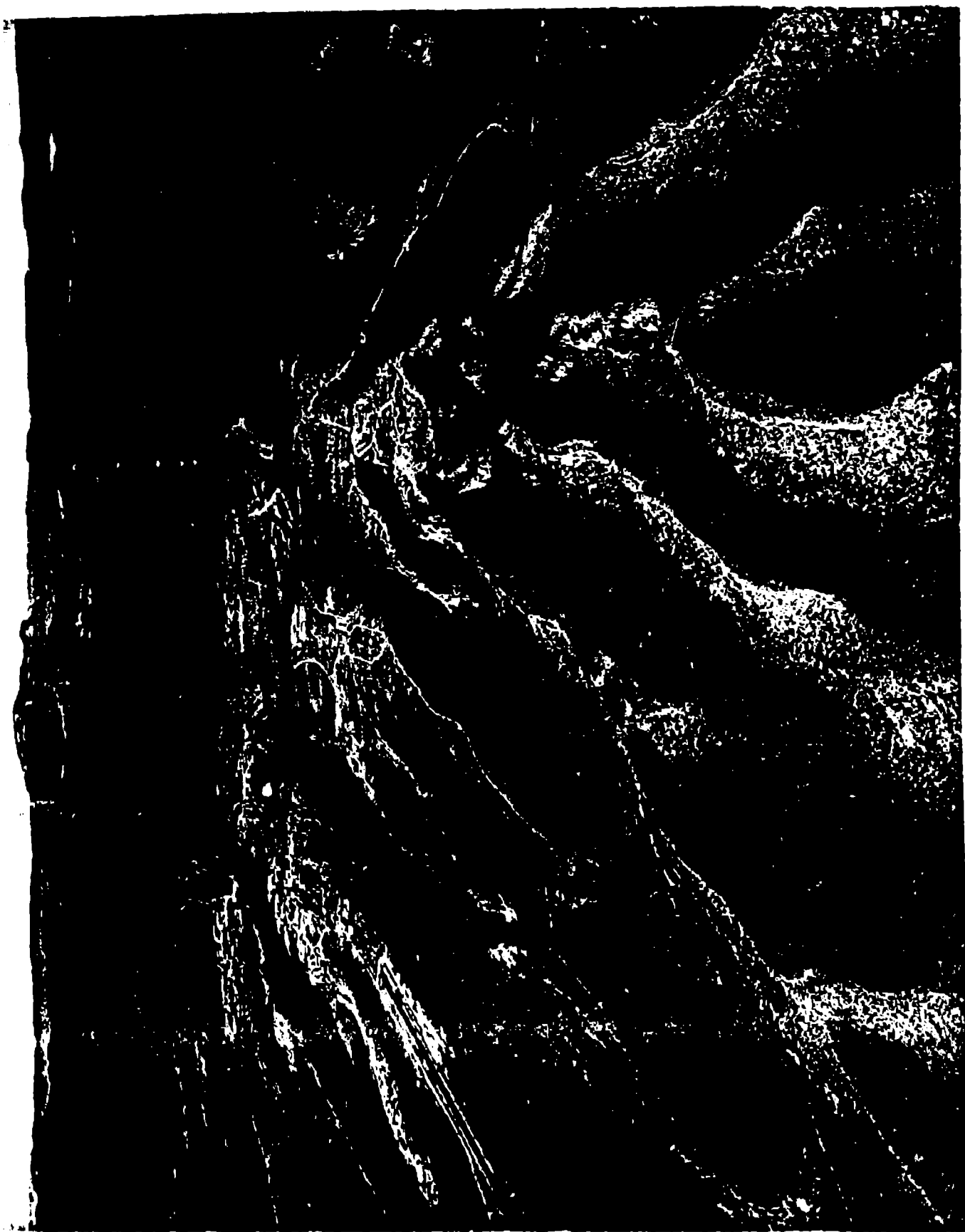
layers of tuff overlying pumice, basalt and other forms of volcanic activity. Runoff from the mountains has eroded deep canyons as it has worked its way to the Rio Grande and the topography of the county consists of a series of "finger" mesas extending to the east. Figure 1 illustrates the location of the county within the state; Fig. 2 provides a picture of the mesa-canyon formations and shows that most of the development is on the mesa tops.

The elevation (2200 m) of the town proper assures a moderate climate with about 0.46 m annual rainfall, much different than weather conditions in central and southern New Mexico. None of the streams in the canyons extending through the county are perennial so their use for adequate water supply or waste dilution is not possible. Wells over 300 m deep supply both the community and the laboratory with water.

The LASL programs at this time are about equally divided between nuclear weapons studies and research and development in energy, radiobiology, laser technology, safeguards, waste management and other areas. Industrial wastes from these programs vary widely as studies change, but generally they can be described as containing ^{238}Pu and ^{239}Pu as the main radioactive alpha isotopes; minor amounts of ^{241}Am , ^{235}U and ^{238}U ; varying low concentrations of radiostrontium, cesium and mixed fission products; a wide array of common and exotic chemicals with the more difficult to remove being

STATE OF NEW MEXICO

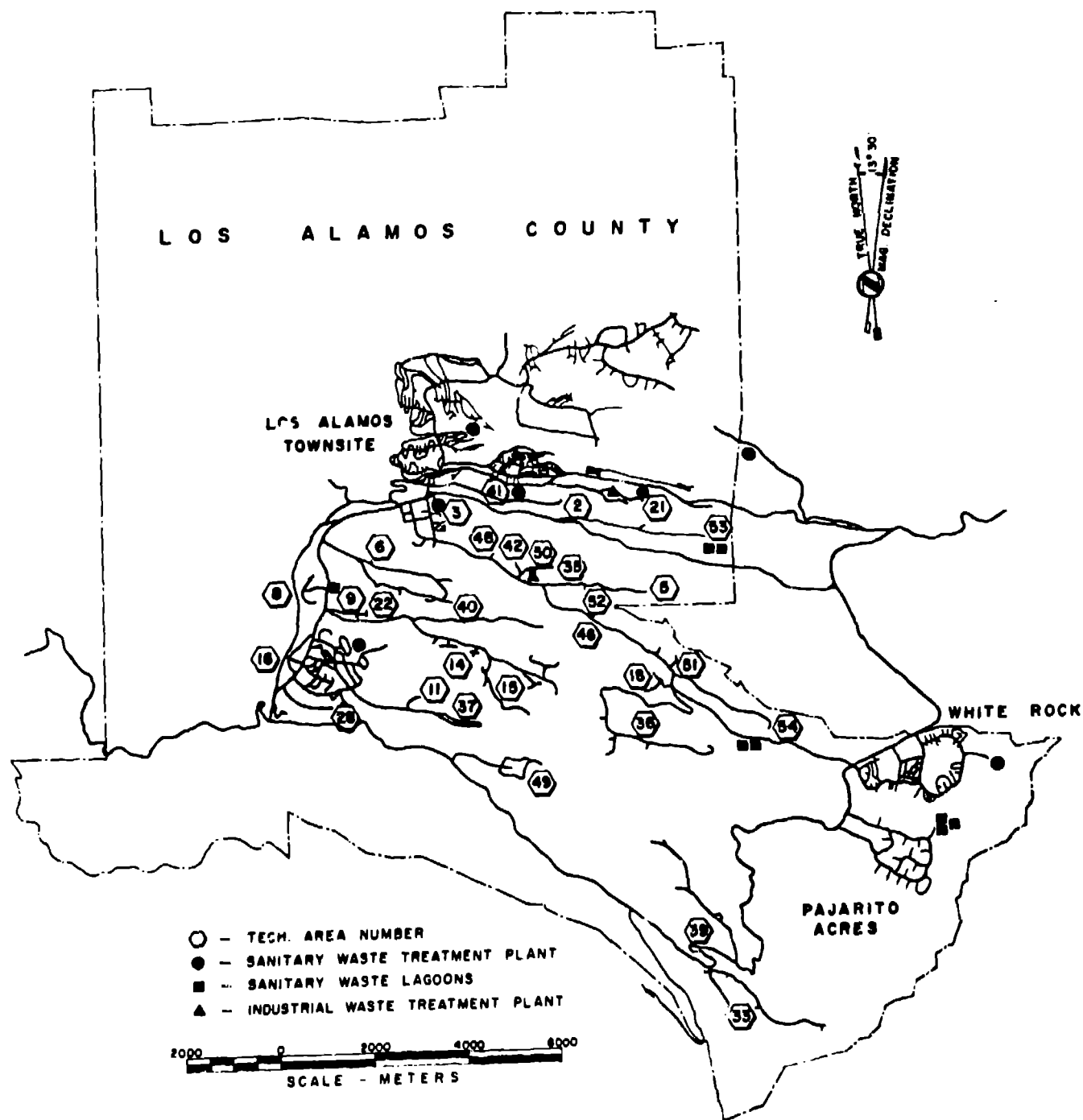




nitrates and fluorides. A high percentage of these wastes are collected in separate industrial waste sewer systems for delivery to either of two waste treatment plants.

A philosophy of waste management was established in the early development of LASL which dictated maximum service to the scientific staff by provision of sewer service to the laboratories with minimum restrictions on discharges. The emphasis now has shifted to water conservation and separation of those wastes with no potential for radioactive or chemical contamination. This has resulted in a gradual decrease in flow volume since 1968. Because of topography, wide separation of project areas and program differences, two separate treatment facilities were provided; one servicing a plutonium processing facility and the other serving a number of separate study centers or technical areas. The original treatment plants were pilot facilities; both have been replaced. The plant serving the plutonium processing facility is known as facility TA-21-257, "TA" representing "Technical Area." The other, or more central plant, is known as the TA-50 facility. Figure 3 indicates the respective locations of the two facilities among the other LASL technical areas.

The group responsible for operation of the waste treatment facilities at LASL is the Waste Management Group, a part of the Health Research Division. The Group's responsibilities extend to all aspects of liquid waste management



from collection to treatment and disposal. In addition, the Waste Management Group controls the collection and disposal of solid wastes which include nonradioactive chemical and radioactive solid materials. These wastes are buried/stored in designated, controlled areas in Los Alamos County. The Group also conducts research and development studies on solid waste volume reduction, the first phase of which is controlled air incineration.

II. WASTE CHARACTERIZATION

From the late 1940's to date, the main radioactive contaminant in the waste has been plutonium. Treatment processes in use were designed to concentrate plutonium in a chemical sludge, to remove this sludge from the stream, to dewater it as much as practicable and then to bury it on site. Up until the early 1970's, the isotope ^{239}Pu was predominant. As programs have changed, much more of the plutonium activity has been due to ^{238}Pu . For CY1976, ^{238}Pu constituted 88% of the plutonium in wastes to TA-50 and 33% of the plutonium wastes to TA-21-257. Other predominant alpha emitters in wastes to both plants include uranium 235 and 238 and americium 241. Strontium 89 and 90 are beta-emitters of concern and cesium 137 is the principal gamma emitter. Because of their low levels in the raw wastes, only a minor amount of effort has been directed specifically toward strontium or cesium removal. This position is changing, however, as environmental monitoring in waste discharge

areas detects rising cesium concentrations in the receiving stream alluvia.

The weak beta emitter, tritium, has not been a problem to date, but this condition also appears to be changing. A number of new facilities which will utilize large amounts of tritium are being constructed and existing accelerators, the meson physics unit in particular, have increasingly larger amounts of tritiated water in waste discharges as power levels are increased.

Tables 1 and 2 provide some general data on the chemical characteristics of wastes to the TA-50 and TA-21-257 plants. Problem areas are nitrates, fluorides, total solids, and, in some instances, certain heavy metals. In the treated wastes, the total solids concentration is due almost entirely to dissolved solids as the filtration step removes settleable and suspended matter.

III. WASTE TREATMENT METHODS

Early studies (Ch51), (Ru52) indicated that chemical treatment utilizing a ferric hydroxide precipitation at a high pH was very effective in sweeping the particulate alpha activity from waste solutions; both Los Alamos plants use this process. Using lime to raise the pH, the floc blanket is increased by the formation of calcium carbonate and removal efficiencies in excess of 99% are achieved. Though various iron salts have been tried, ferric sulphate has been used most commonly because of lower cost, ease of handling

TABLE 1
Chemical Characteristics of Waste
TA-50 Plant
Annual Average Values, mg/l

	1974		1975		1976	
	Raw	Treated	Raw	Treated	Raw	Treated
Total Solids	585	1540	690	1200	490	1520
Na	150	430	140	470	120	450
Ca	15	43	14	21	24	21
NO ₃ -N	12	66	16	102	25	90
F	5.0	2.6	2.6	1.8	5.8	4.5
PO ₄	4.9	.3	4.9	.7	3.3	1.2
Cd	.015	0.02	.011	.002	.012	<.001
Total Cr	.10	0.02	.18	.07	.30	.12
Hg	.049	0.003	.049	.003	.12	<.005
Pb	.07	0.05	.14	.03	.17	<.001

TABLE 2
Chemical Characteristics of Waste
TA-21-257 Plant

Annual Average Values, mg/l

	1974		1975		1976	
	Raw	Treated	Raw	Treated	Raw	Treated
Total Solids	3590	3470	4930	4640	5290	4570
Na	1060	960	1310	1260	1070	1010
Ca	24	16	12	6	29	13
NO ₃ -N	274	260	352	315	476	420
F	44	42	29	9	50	32
PO ₄	--	0.6	1.3	1.9	3.8	0.8
Cd	.033	0.040	<.004	<.003	.060	<.003
Total Cr	.037	0.030	.042	.057	.061	.038
Hg	.017	0.001	.016	<.001	.029	<.001
Pb	.05	0.04	.04	<.02	.08	<.02

and somewhat greater acceptability of sulfates in the waste stream. Tests (Dr76) were made with magnesium sulfate as the coagulant and in many ways it was equally effective. It has the advantage of forming a somewhat gelatinous floc which is much more efficient in alpha activity removal during periods when the wastes contain soaps or chelating agents. However, for continuous use, it has the drawback of producing a very light floc which does not settle readily and a more voluminous magnesium hydroxide sludge which is difficult to dewater.

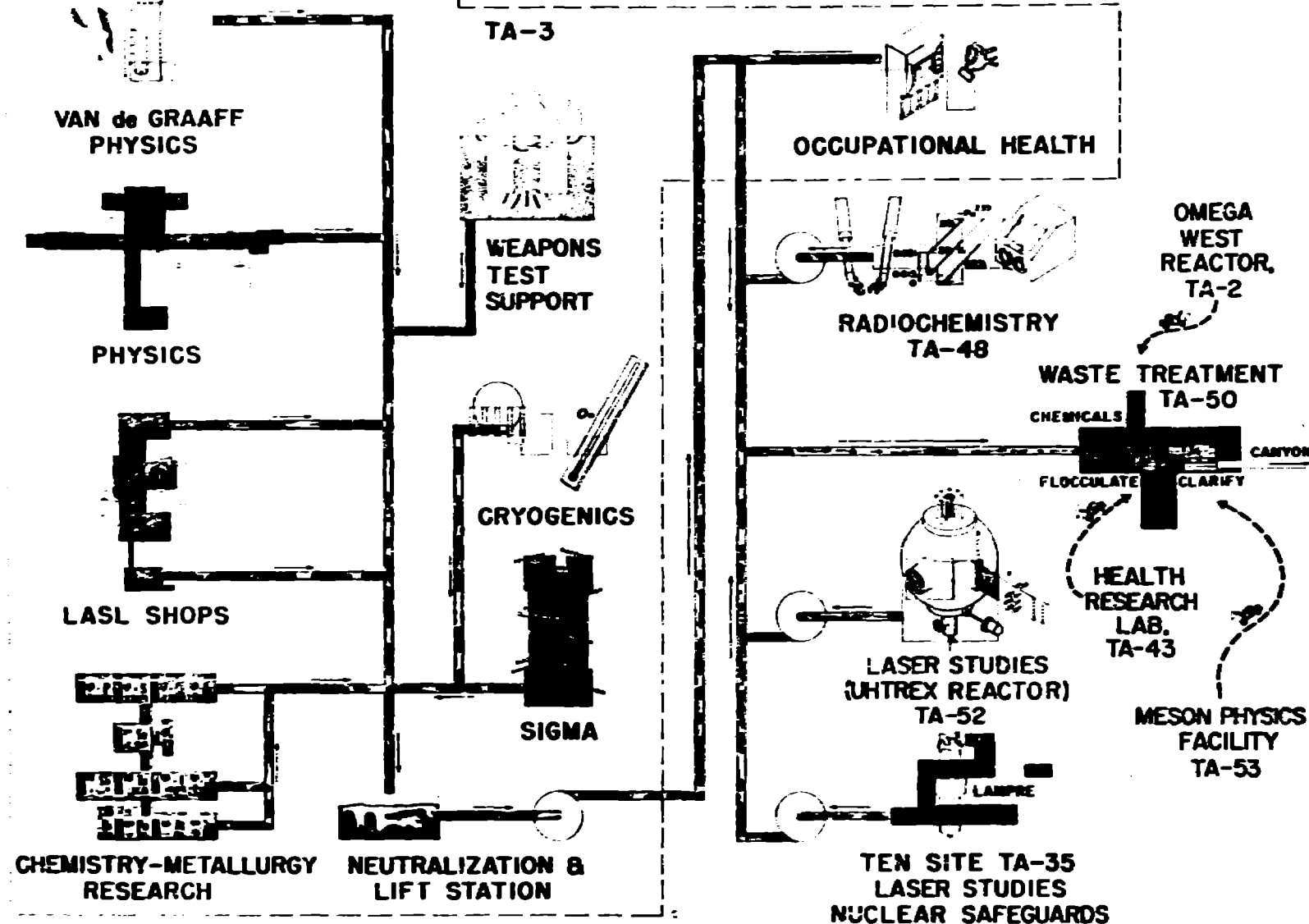
The high pH ferric hydroxide precipitation is very inefficient for strontium or cesium removal; cesium removal can be accomplished in chemical treatment through the use of potassium ferrocyanide and nickel sulphate. Other methods include use of synthetic cation exchange resins, synthetic zeolites (W174, W1176) and natural zeolites such as clinoptilolite. At the TA-50 plant, high capacity cation exchange resins are a part of the process. Operating in the hydrogen phase, they efficiently remove both cesium and strontium. However, as column loading continues, divalent ions such as calcium (water hardness) replace the cesium on the resin beads and overall cesium removal is very poor. Studies are being conducted to determine cycle length for cesium breakthrough and to investigate use of certain sulfonic acid exchangers (phenolic) which are reported (W1175) to be somewhat selective for cesium.

IV. WASTE COLLECTION SYSTEMS

As mentioned previously, the primary means of transporting wastes from generator to treatment plant is through separate sets of industrial waste sewers. For the TA-21-257 plant, these sewers are of cast iron and stainless steel construction, are single envelope only and are installed in areas not readily accessible to the general public. For the TA-50 plant, the sewers are of vitrified clay tile, cast iron and polyethylene construction, are of single envelope only and a high percentage of the length passes through areas which are open to the general public. Figure 4 is a schematic of the sewer system which delivers wastes to the TA-50 facility. The primary sources of radioactivity are the chemistry-metallurgy research building and the radio-chemistry site.

As may be noted in Fig. 4, a small amount of wastes are collected by tank truck, in this case 3800-liter Dempster Dumpster units, and hauled to TA-50. The wastes have very low-levels of radioactivity, primarily beta-gamma, and originate at an accelerator site, a small research reactor and at a health research laboratory. In numerous instances, small waste quantities packaged in containers ranging in size from about 5 m³ to 200 liters are collected by truck for delivery to TA-50 or TA-21-257.

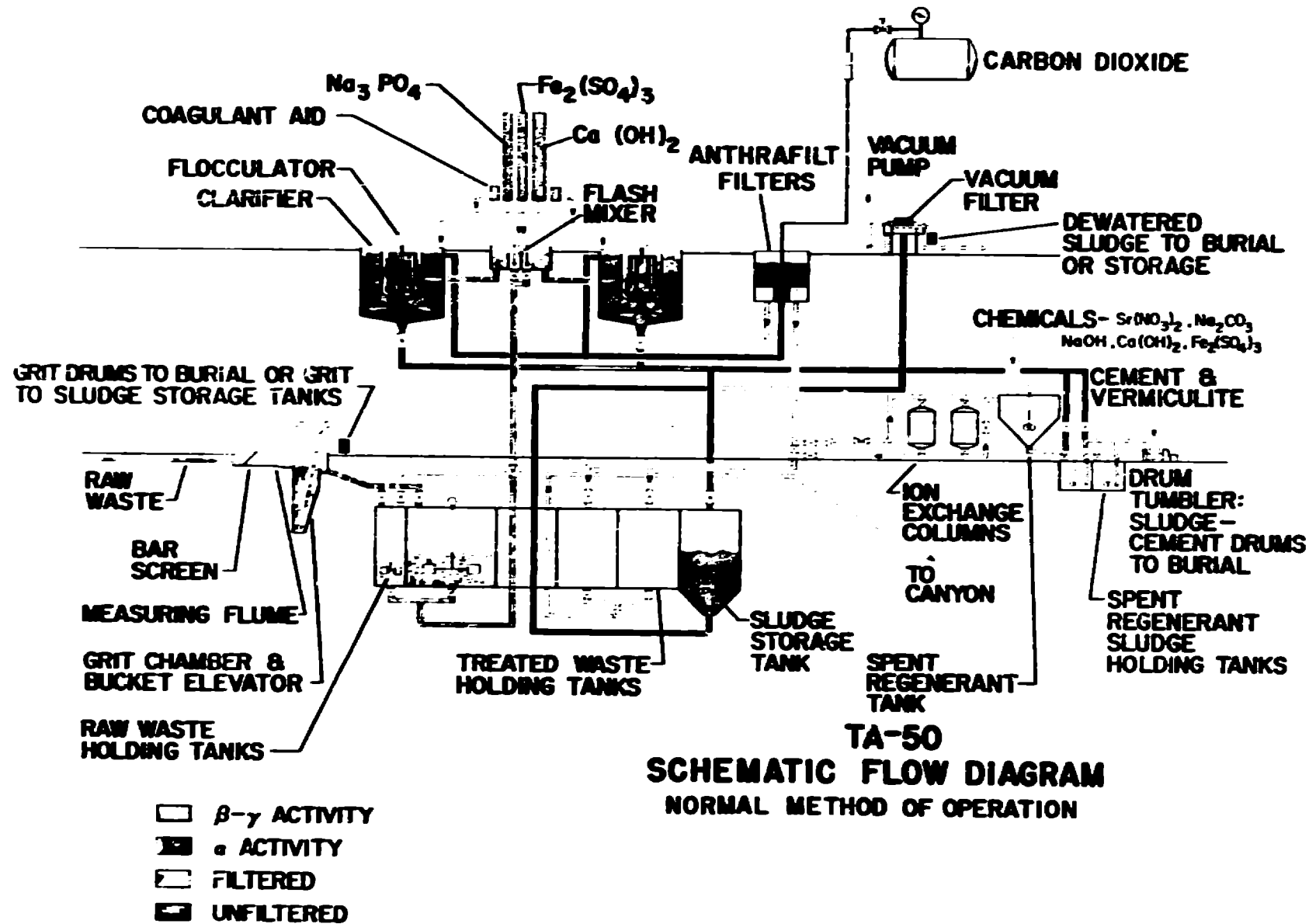
WASTE COLLECTION SYSTEM SERVING TA-50



V. TA-50 WASTE TREATMENT PLANT

A schematic of this facility is shown in Fig. 5 and views of the operating floor are provided in Figs. 6 and 7. The plant is designed to treat wastes chemically at a rate of about 950 liters per minute over a normal operating schedule of eight hours per day, five days per week. 380,000 liters of storage capacity are adequate to permit the single shift operation.

Wastes pumped from storage are delivered to flash mixers where about 300 mg/l of lime, 100 mg/l of ferric sulfate, about 200 mg/l of trisodium phosphate and recirculated sludge are added. The mixture flows through flocculator (~ 30 min), sedimentation tank (2.5 hrs) and gravity filters to pumps which send it through the ion exchange columns to treated-waste storage. The settling tank effluent is recarbonated using liquid carbon dioxide to convert excess lime to the bicarbonate and prevent it from cementing filter media. When a storage tank is filled with treated waste, alpha radioactivity of a composite sample is determined. If this activity level, assumed to be due to plutonium, is less than 50% of the RCG as listed in Table II of ERDA Manual Chapter 0524 Appendix ($5 \times 10^{-6} \mu\text{Ci/cc}$), the liquid is pumped to a canyon to the north of the plant. If the treated waste sample indicates a higher level of activity, the entire tank contents are recycled to the raw







waste storage tank for retreatment. Some operating data are provided in Table 3.

Filters are backwashed with treated waste water when head loss dictates and washwater is returned to raw waste storage.

Sludge is drained weekly to sludge storage where it can be concentrated to about a 10% solids level by decanting. From storage, sludge is pumped to a pre-coat type vacuum filter where it is dewatered to about 30-40% solids. If transuranic (TRU) activity level of the sludge is ≤ 10 nCi/g, the dewatered sludge is collected in a 5-mil polyethylene-lined 200-liter fiber drum or 215-liter steel drum and buried at a designated, controlled site within the LASL boundary. If TRU activity is > 10 nCi/g, the sludge is packaged in 90-mil, polyethylene-lined, 215-liter, DOT 17C steel drums and stored retrievably at the same designated waste management site. In CY1976, the dewatered sludge averaged 3 to 4 nCi/g; 630 fiber drums and 83 steel drums (nonretrievable) were sent to burial.

The ion exchange resins are regenerated with 6 N nitric acid. The spent regenerant is collected in a special tank and treated chemically to concentrate the radioactivity. This chemical sludge is usually drained to the sludge storage tank, but it can be pumped directly to the vacuum filter or it can be mixed with cement in a separate operation.

TABLE 3
OPERATING DATA
TA-50 TREATMENT PLANT

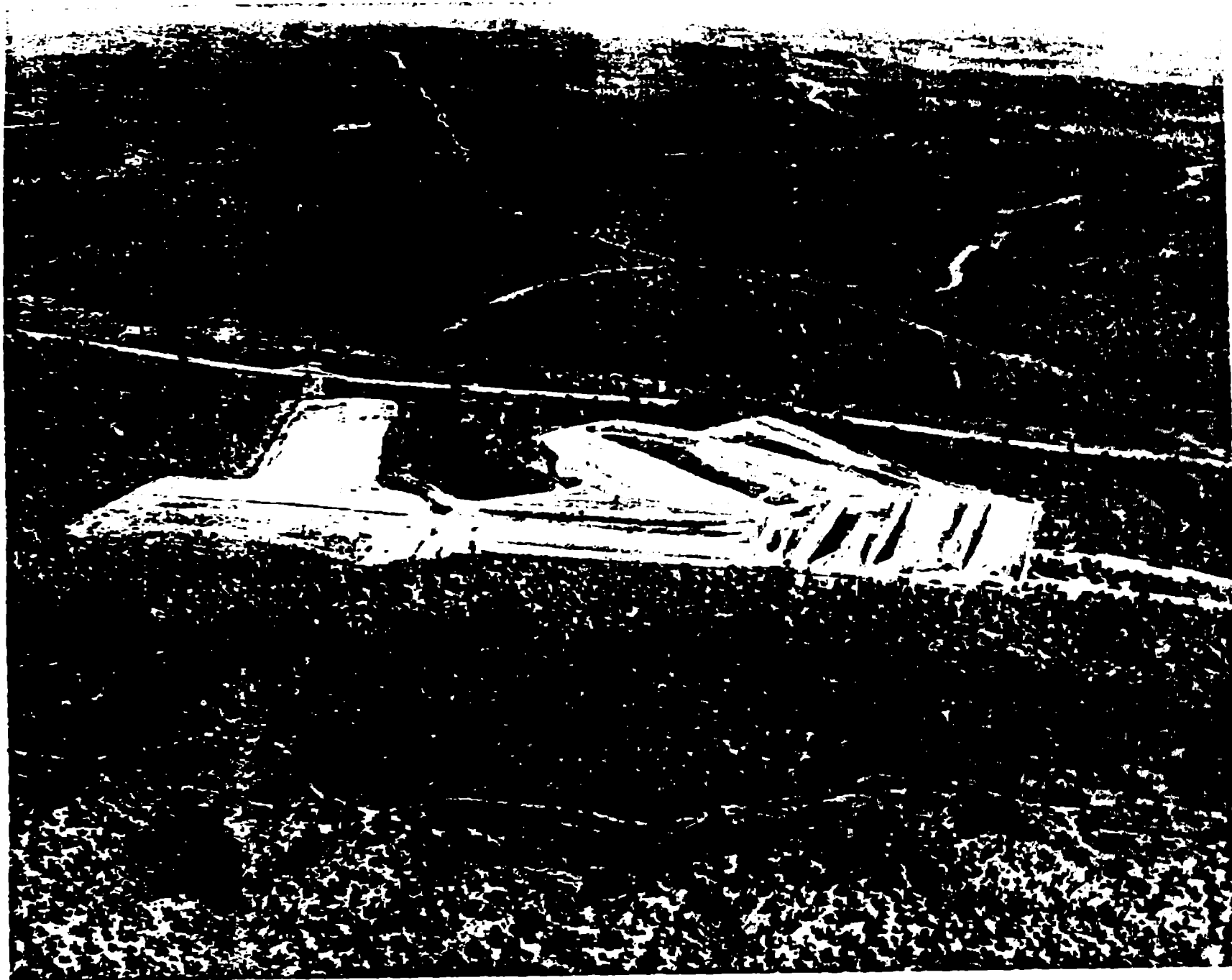
Year	Vol. of Waste Discharged, 10 ⁶ Liters	Ave. α Activity, $\mu\text{Ci/cc}$		Decon. Factor	Vol. of Dewatered Sludge, Liters	Ave. Volume Reduction Factor
		Raw Waste	Treated Waste			
1971	46.2	8.60×10^{-5}	2.62×10^{-7}	325	156,500	424
1972	59.4	1.40×10^{-4}	2.43×10^{-7}	683	242,440	326
1973	52.4	1.72×10^{-4}	2.85×10^{-7}	621	185,420	283
1974	40.6	1.33×10^{-4}	3.38×10^{-7}	418	102,300	397
1975	39.7	1.94×10^{-4}	5.00×10^{-7}	382	153,330	259
1976	39.9	8.56×10^{-5}	2.51×10^{-7}	434	136,810	292

The radioactive solid waste burial/disposal site to which the dewatered sludge is transferred is located approximately four miles east of the TA-50 treatment plant.

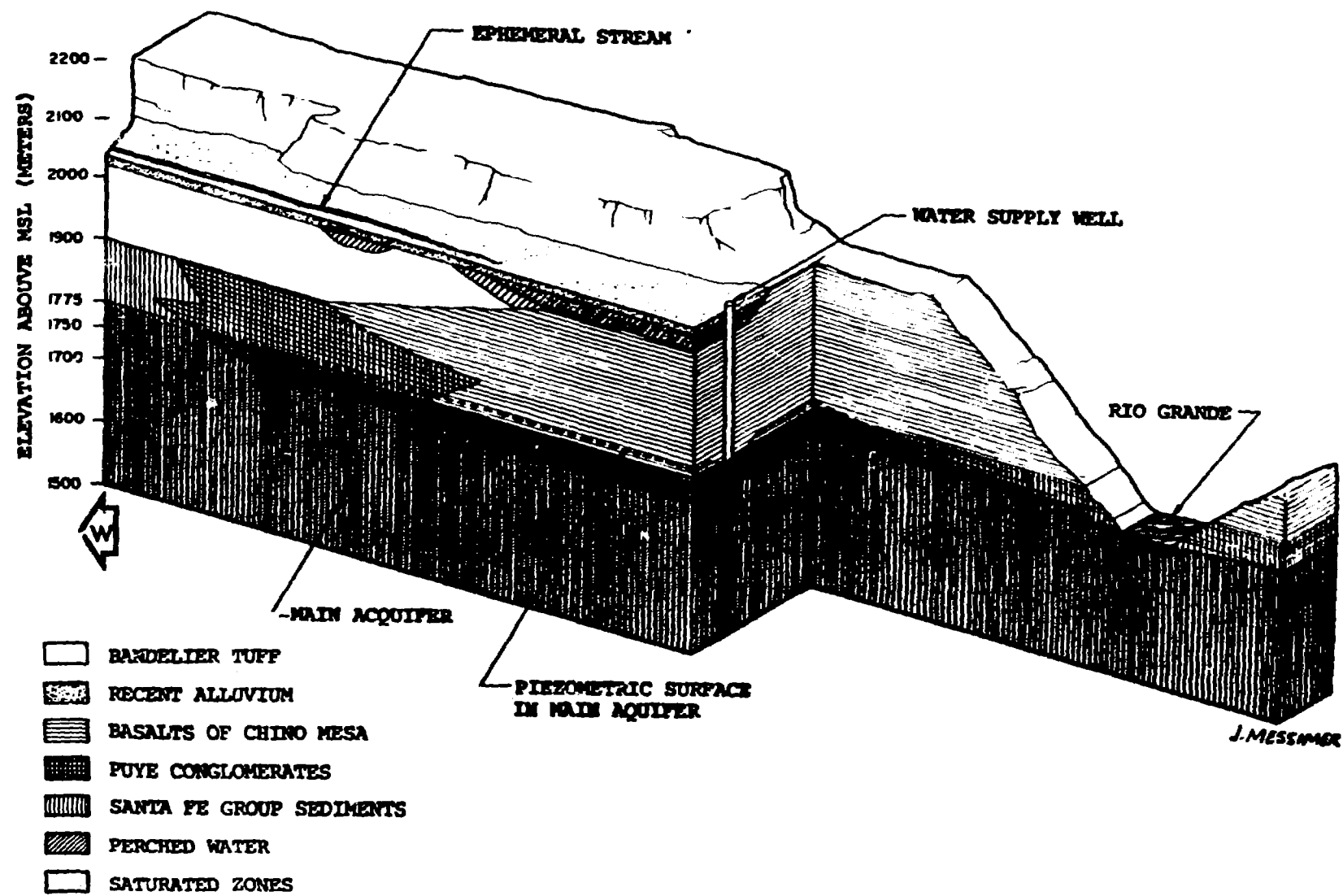
(Fig. 8) The rainfall, evaporation and geology are such that infiltration of water is very low and the only available water table is still several hundred meters below the site. (Fig. 9) The nonretrievable radioactive waste drums from the treatment plants and miscellaneous nonretrievable trash from other sites are placed in pits excavated directly in the tuff. (Fig. 10) As drums and other wastes are added to the pit, they are continuously covered; when the pit is completely filled, the tuff is mounded over the top to provide a protective layer several meters thick. Retrievable drums are placed in 20-year storage. (Fig. 11)

VI. TA-21-257 WASTE TREATMENT PLANT

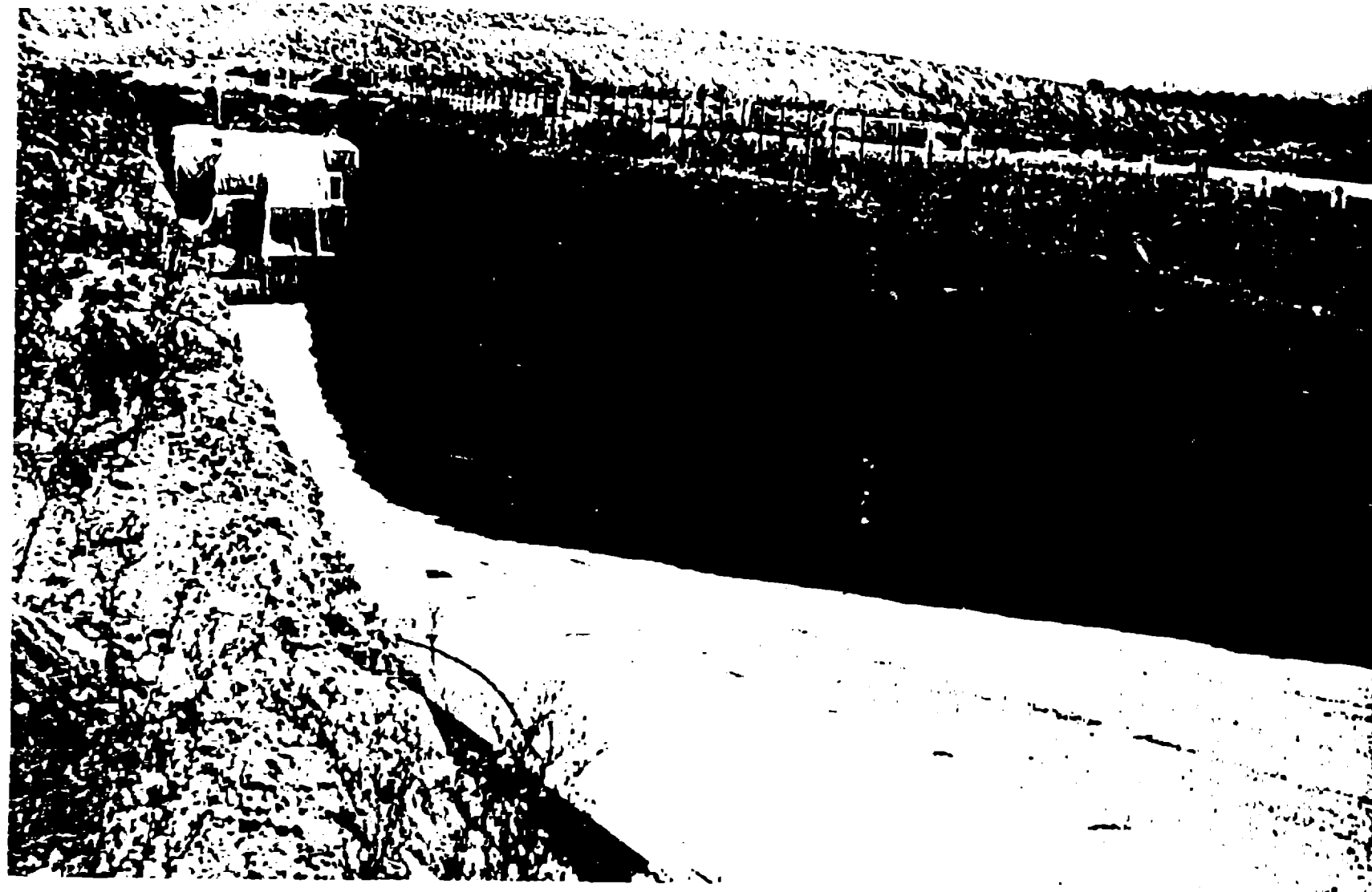
Figure 12 provides a schematic of the "257" plant. References (Em70) and (Em71) provide a detailed history of plant development in this area. The process is identical to that at TA-50 with the exceptions that no ion exchange is provided and the chemical sludge is increased rather than decreased in volume by the addition of cement. The "257" plant is designed to treat at the rate of about 475 gpm through flash mixer, flocculator, sedimentation tank and filter. In this case, flocculator and clarifier are separate units and the filter is a pressure filter rather than a gravity unit as at TA-50. Recarbonation of

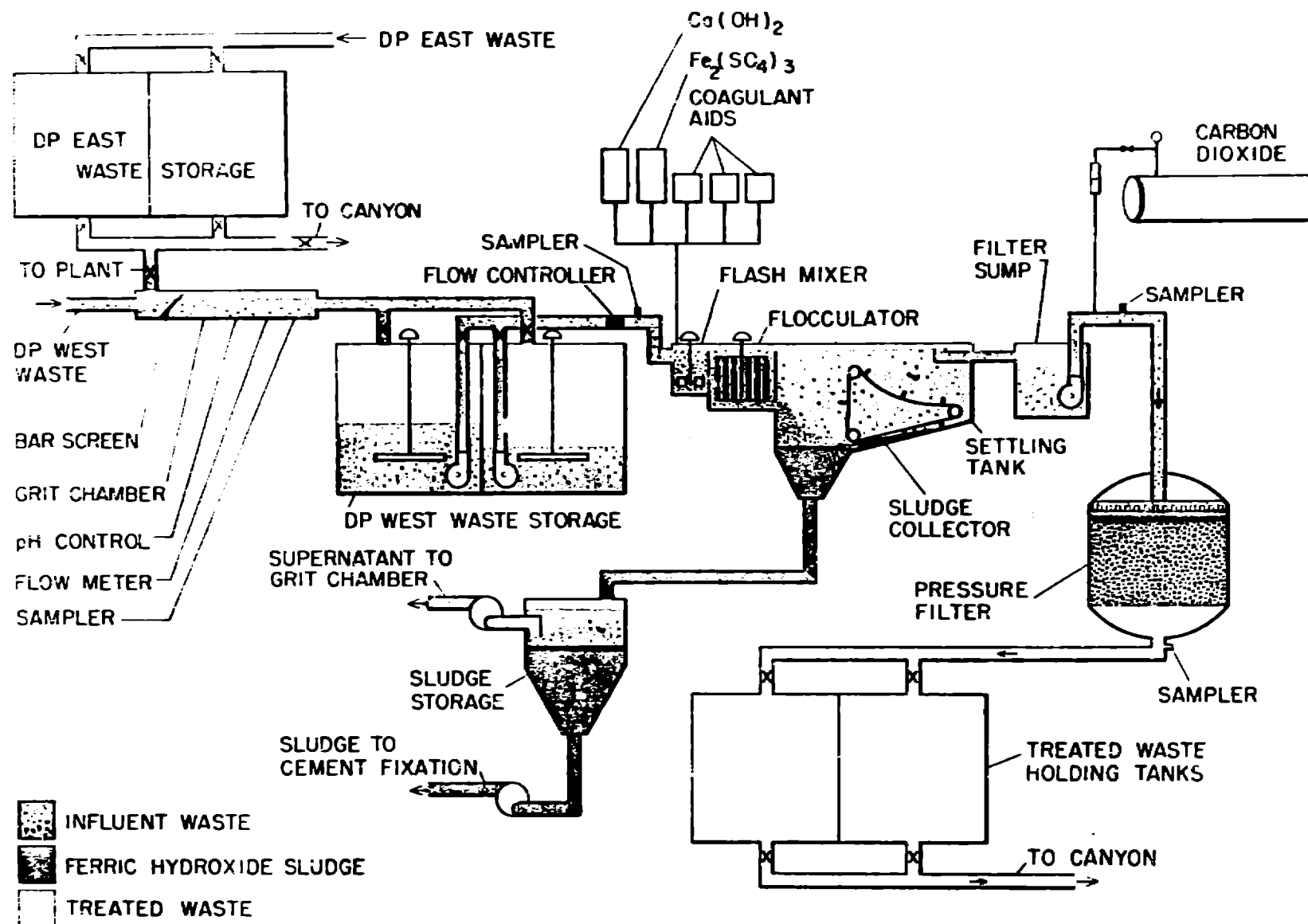


CONCEPTUAL HYDROLOGICAL CROSS SECTION OF THE LOS ALAMOS AREA









LIQUID WASTE TREATMENT PLANT
LOS ALAMOS SCIENTIFIC LABORATORY

settling tank effluent is accomplished through the use of dry ice. Plant performance has been very satisfactory as is evidenced by the data of Table 4. It is noted from the table that annual average decontamination factors for this plant are much more erratic than for the TA-50 plant, but the average of these factors over the last five years is much higher for the "257" facility. No serious attempts to pinpoint the reasons for the differences in decon factors have been made, but it seems likely that waste composition (e.g., concentration of chelates) and settling tank short-circuiting are involved.

At the "257" plant, the chemical sludge is mixed with cement in a pug mill, an open horizontal mixer as used in the ceramics industry. As shown in Fig. 13, waste and cement are introduced at one end of the mill, then mixed as they are conveyed to the other end by a series of paddles. The system was originally installed to "fix" a special batch waste containing mg/l quantities of ^{241}Am and ^{239}Pu in a cement paste. This liquid was deemed to be too radioactive to treat through the open chemical precipitation plant in the volumes, about 6500 liters per month, being received. The continuous treatment pug mill system which was installed has a capacity far greater than that needed to process the americum waste only. It was adapted, therefore, to fix other batch wastes plus the plant chemical sludges in cement.

TABLE 4
OPERATING DATA
TA-21-257 TREATMENT PLANT

Year	Vol./Waste Discharged 10 ⁶ Liters	Ave. Activity, $\mu\text{Ci/cc}$		Decon. Factor	Effluent Activity Total mCi			
		Raw Waste	Treated Waste		Pu	²⁴¹ Am	⁸⁹⁻⁹⁰ Sr	³ H
1971	9.91	5.74×10^{-5}	0.8×10^{-7}	814	.71	N.A.	1.02	3000
1972	8.99	5.05×10^{-5}	1.1×10^{-7}	585	1.30	N.A.	1.67	3650
1973	5.44	1.07×10^{-4}	1.7×10^{-7}	804	.41	.14	.68	1488
1974	4.56	2.07×10^{-4}	1.1×10^{-7}	2309	.21	.22	.44	648
1975	5.75	3.16×10^{-4}	3.6×10^{-7}	662	.95	.33	.70	9100
1976	4.70	1.20×10^{-4}	1.4×10^{-7}	1133	.39	.13	.50	870

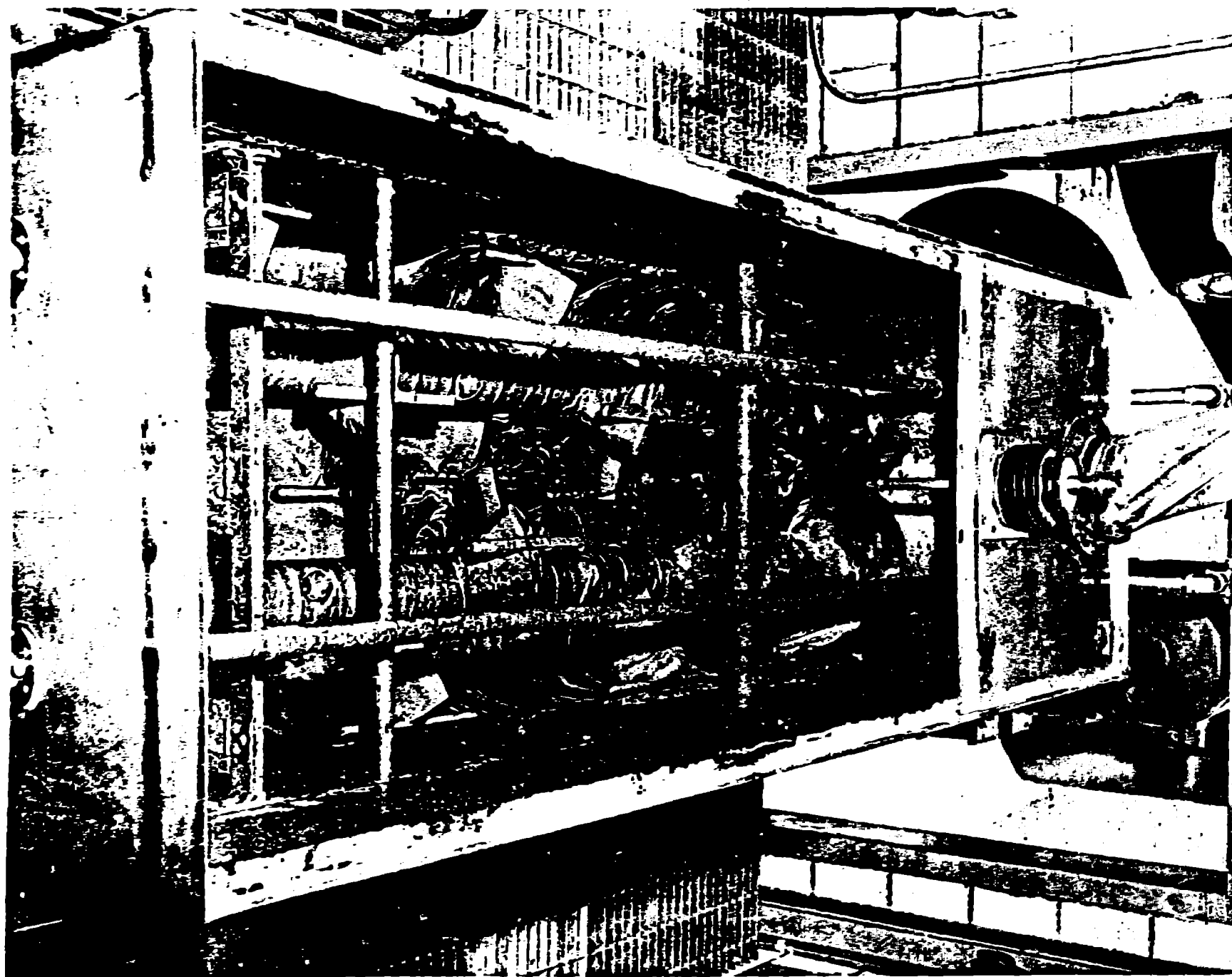


Table 5 provides some data on the last several years of operation. A schematic of the system is shown in Fig. 14.

When pug mill operations began in 1968, and for several years thereafter, all cement paste was pumped to 2.44 m deep shafts which were drilled in the volcanic tuff just west of the treatment plant. Upon promulgation by ERDA of Manual Chapter 0511 establishing retrievability requirements, pastes above the retrievability level of 10 nCi/g were discharged to packages consisting of 6.1 m lengths of 0.76 m diameter galvanized, welded corrugated metal pipe. A "cold" concrete plug about 0.3 m thick is poured at bottom and top to provide a package which has a contamination-free exterior. Table 6 lists information on the shaft operations and Fig. 15 is a view of the pit containing the corrugated metal pipe.

In an attempt to determine the effectiveness of this cement "fixing" operation, two tests are conducted. In the first test, 152 mm diameter by 305 mm long cylinders of cement paste are collected during a run, permitted to cure and develop strength, then are broken in a compressive strength testing apparatus. Curing is conducted with the sample submerged in tap water for a period of about 30 days. Compressive strengths have averaged about 10.34 MPa (1500 psi) with the maximum up to 27.58 MPa (4000 psi). Although the cement paste when disposed to the pits and CMP is not under compression, the tests provide an indication of the

TABLE 5
PUG MILL OPERATION AND DATA

<u>YEAR*</u>	<u>Vol. of Liquid Feed, Liters</u>	<u>Vol. of Cement Paste, Liters</u>	<u>Total radioactivity in Paste, Ci</u>			
			<u>238 Pu</u>	<u>239 Pu</u>	<u>241 AM</u>	<u>MFP</u>
1972	313310	533450	3.7	8.2	N.A.	.5
1973	320460	547540	10.7	64.9	N.A.	1.4
1974	262870	521740	24.1	90.1	685.4	1.1
1975	260870	457360	8.9	29.2	784.8	6.5
1976	208580	290,820	15.0	29.1	704.0	1.7

* 1975 and 1976 data is for material in both retrievable and nonretrievable shafts.

PUG MILL SYSTEM

Continuous Mixing of Waste & Cement

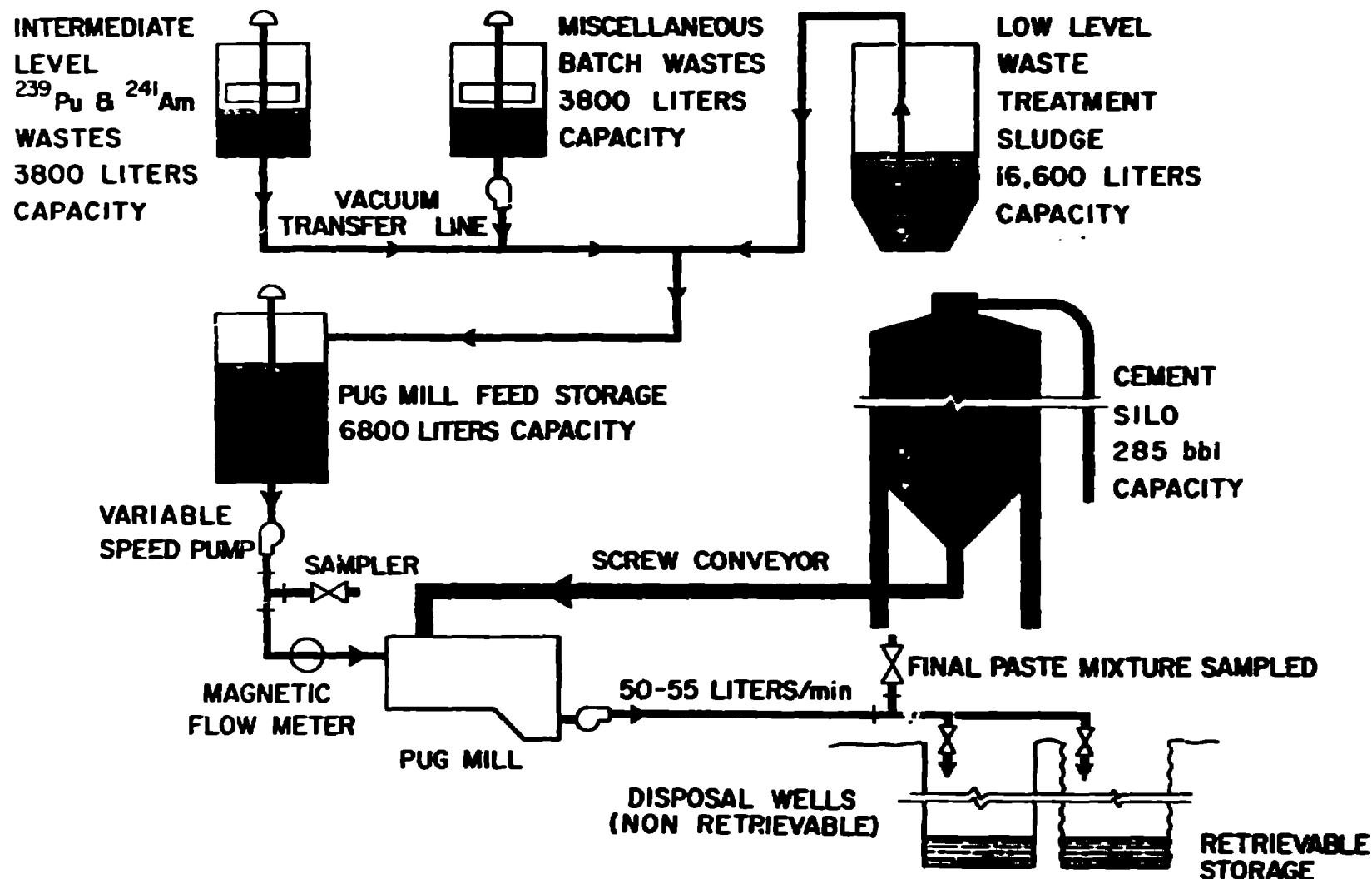
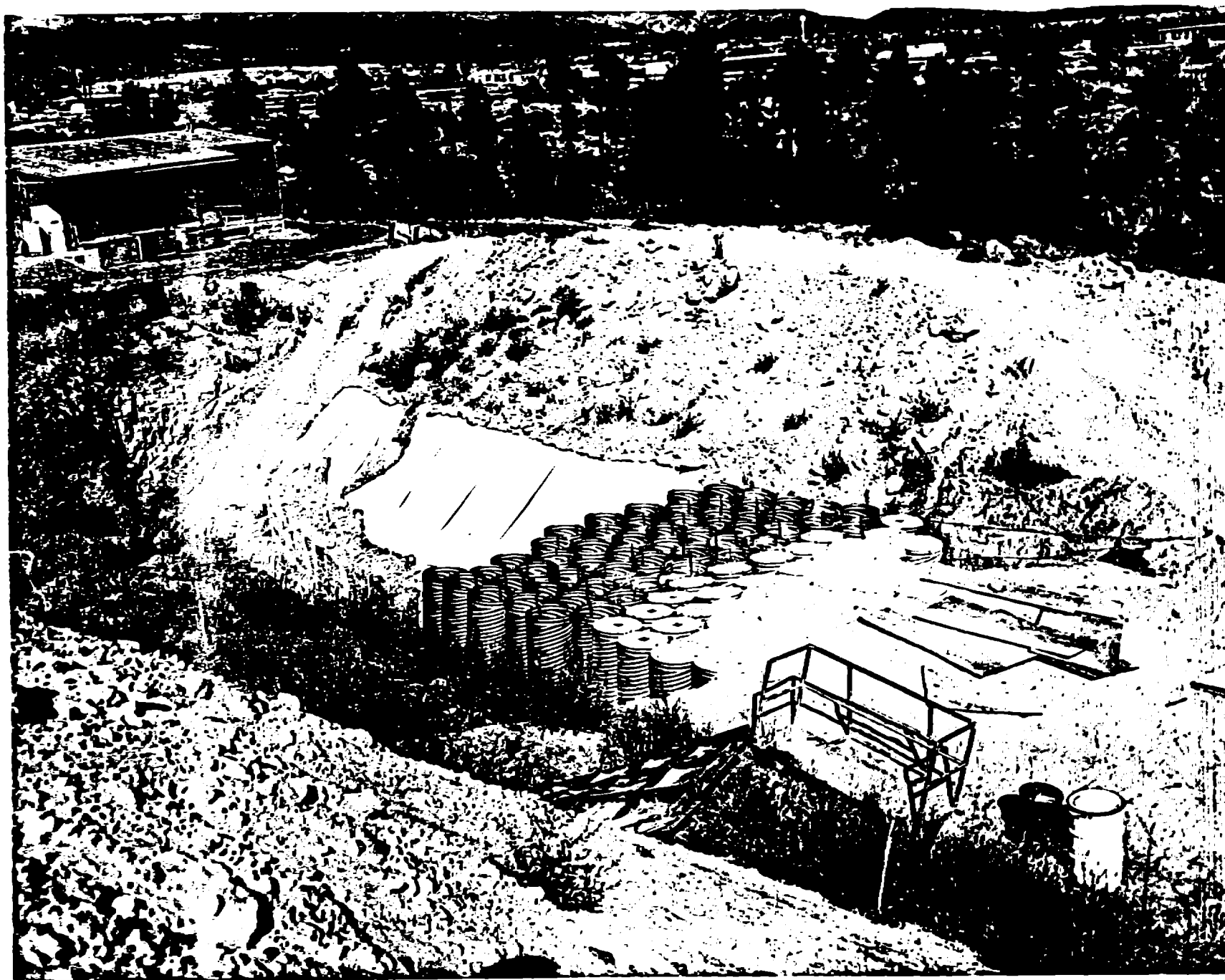


TABLE 6
PIT DATA
PUG MILL OPERATIONS
TA-21-257

YEAR	No. 2.4m Shafts Filled	Avg. Vol. Paste per Shaft, liters	No. 1.83 m Shafts, Filled (1),(2)	Avg. Vol. Paste per Shaft, liters	No. CMP Filled	Average Vol. paste per CMP (excl. caps)
1972	7	76,600	--	--	--	--
1973	9	69,300	--	--	--	--
1974	6	77,200	--	--	--	--
1975	5	54,700	4	44,000	2	2160
1976	-	--	3	47,900	56	2440

(1) Not all pits were 20 m deep

(2) 1.83 m diam. pits were drilled in the space between the rows of 2.1 m pits.



quality of the mix. From this information, it is possible to determine which waste solutions adversely affect setting.

In the second test, small (~ 60 cc) paste samples are collected in jars, permitted to set for a day, then are removed from the container and immersed in tap water. The immersion water is counted and replaced after one day, then one week, then one month, then annually. Percentages of radioactivity leached, as an example, are given in Table 7. Alpha activity is very tightly bound but nuclides such as ^{137}Cs (γ) and ^3H (β) are easily leached from the paste.

VII. FUTURE IMPROVEMENTS

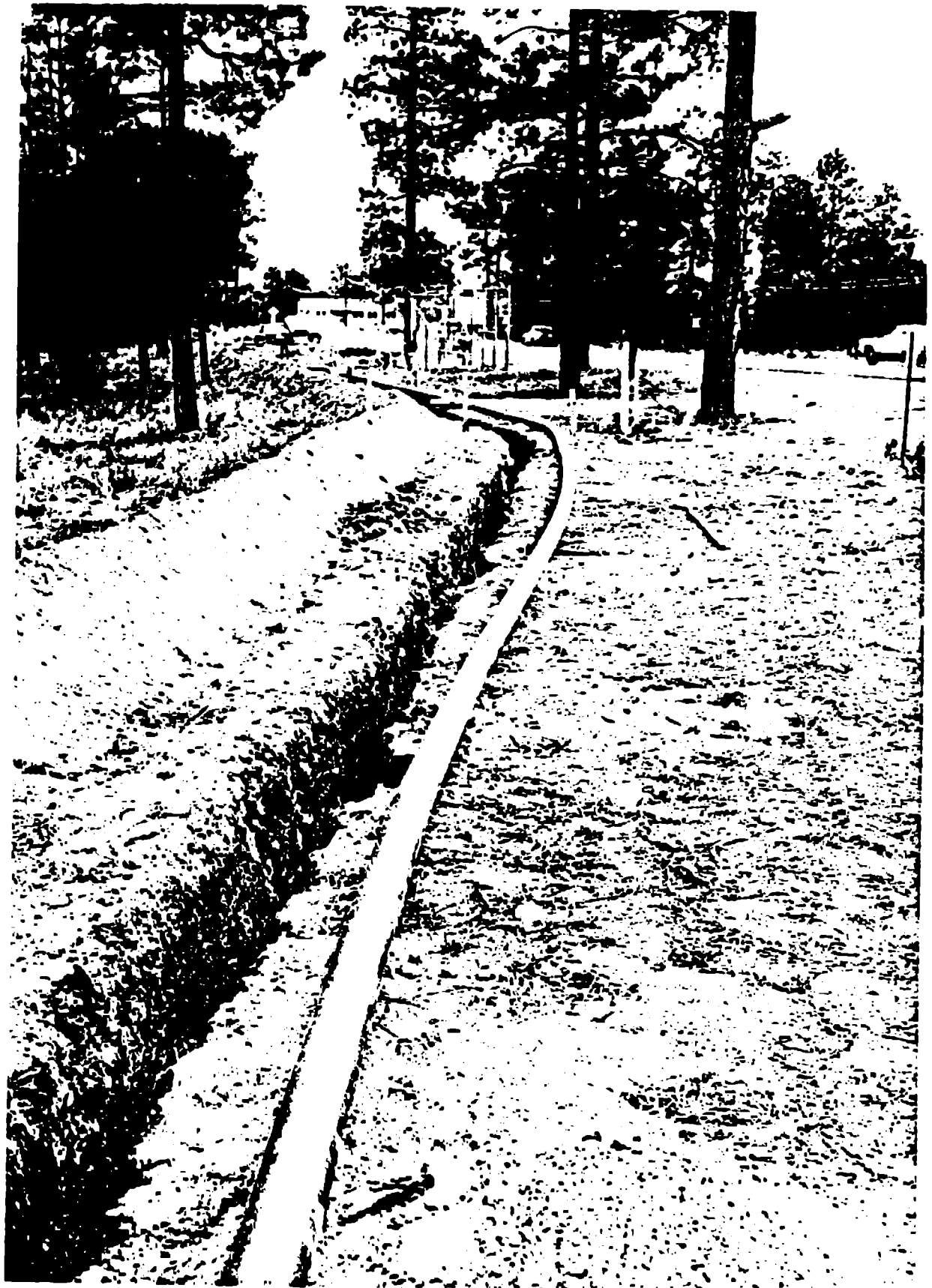
A. Sewer Systems

In 1974, a thorough review of the existing sewer system was initiated. It was decided that vitrified clay pipe (VCP) in its relatively short lengths with joints at frequent intervals was no longer satisfactory for transfer of radioactive wastes, and in one section, several thousand feet of the VCP were replaced by high density welded polyethylene pipe. Figures 16 and 17 were taken during the installation. This installation, however, was a temporary measure which addressed only part of the system. Therefore, plans and design studies are underway to provide for:

- construction of a complete, double-encased, monitored sewer system
- removal of all abandoned radioactive waste sewer lines and any contaminated soils in the trenches

TABLE 7
LEACH TEST RESULTS
CEMENT PASTE SAMPLES

<u>Date Immersed Initially</u>	<u>Total α Activity, Ci in Sample</u>	<u>Total Time Immersed, Yrs.</u>	<u>% α Activ- ity Leached</u>
10/27/61	2.2×10^{-2} Ci	14	.00003
10/23/63	6.9×10^{-3} Ci	13	.000009
5/13/65	1.2×10^{-1} Ci	11	.000009
7/14/67	1.1×10^{-1} Ci	9	.0000004
2/21/69	4.6×10^{-4} Ci	6	.0010
3/02/70	1.6×10^{-4} Ci	6	.0005
4/20/71	4.4×10^{-4} Ci	5	.0012
4/24/72	3.4×10^{-4} Ci	4	.0020
4/10/73	6.2×10^{-4} Ci	3	.0014
4/08/74	1.4×10^{-4} Ci	2	.0004





- cleaning, resealing and monitoring of existing rad waste storage tanks
- installation of an electronic monitoring-control system utilizing multiplexing

B. Waste Treatment Plants

Presidential and AEC/ERDA orders and directives require contractor facilities such as the LASL to comply with federal and state pollutant levels in effluents and to discharge the minimum practicable amount of radioactivity. The Environmental Protection Agency, in compliance with the Water Pollution Control Act, Amendments of 1972, (PL 92-500), is negotiating with ERDA to establish effluent guidelines for nonradioactive pollutants in the discharges from the two treatment plants. Though discharges to date have contained a very low level of radioactivity, environmental surveillance has detected accumulations of plutonium and cesium in alluvia of the intermittent streams in the receiving canyons.

Because ERDA/LASL believe that the plant effluents must be improved not only from the standpoint of radioactivity but also from the standpoint of certain heavy metals, nitrates and fluorides, planning is also underway for certain plant improvements. In this project, treatment would be revised to eliminate completely any discharges from the treatment plants. This could be accomplished by fossil-fuel fired mechanical evaporation or by solar evaporation of treated effluents in highly engineered ponds. Construction

planning and design funds have been obtained to retain the services of an engineering firm for studies which will pinpoint the optimum process and for preliminary plans (Title I) to implement construction of the selected system.

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